Dilatometric study of $Ni_{2+x}Mn_{1-x}Ga$ under magnetic field

A. N. Vasil'ev,[1] E. I. Estrin,[2] V. V. Khovailo,[3] A. D. Bozhko,[1] R. A. Ischuk,[1] M. Matsumoto,[4] T. Takagi,[3] and J. Tani[3]

[1]*Low Temperature Physics Department, Moscow State University, Moscow 119899, Russia*
[2]*Institute of Physical Metallurgy, Central Research Institute for Ferrous Metallurgy, Moscow 107005, Russia*
[3]*Institute of Fluid Science, Tohoku University, Sendai 980-8577, Japan*
[4]*Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan*

A study of polycrystalline and single crystalline $Ni_{2+x}Mn_{1-x}Ga$ alloys by means of dilatometric and strain gage techniques shows that large strain can be induced in the temperature range of the martensitic transformation by application of an external magnetic field. The biggest strain induced by magnetic field was observed in the samples of chemical compositions where the structural and magnetic transitions couple, i.e. occur at the same temperature.

I. INTRODUCTION

Recently, much activity has been devoted to the development of ferromagnetic shape memory alloys. Various intermetallic compounds have been considered from the viewpoint of a combination of ferromagnetic properties and structural transformations of martensitic type. Among them are the body centered cubic or body centered tetragonal structures of Fe-Ni-Cr, Fe-Ni-C and Fe-Ni-Co-Ti [1-3], and face centered tetragonal structures of Fe-Pt and Fe-Pd [4,5]. Since the magnetic energy is very small compared to the energy stored in chemical bonds, the influence of the magnetic field on the shape memory alloys can be seen only in a limited interval near the temperature of martensite – austenite or austenite – martensite transformation.

The above mentioned were ferrous shape memory alloys, however there exist non-ferrous materials which also exhibit a well pronounced shape memory effect in the ferromagnetic state. The most intensively studied of them is a Heusler type alloy Ni_2MnGa [6-9]. For the stoichiometric composition its Curie temperature is $T_C = 376$ K and the temperature of austenite cubic to martensite tetragonal transformation is $T_m = 202$ K [10]. The partial substitution of Mn by Ni results in the decrease of T_C and in the increase of T_m until these finally merge in the range of compositions near $Ni_{2.19}Mn_{0.81}Ga$ [11]. The sample of this composition and of nearby compositions were the subject of the present study.

II. RESULTS AND DISCUSSION

The measurements were performed on both polycrystalline and single crystalline samples. The sample dimensions were typically $3 \times 3 \times 6$ mm. Studies on polycrystalline samples were done for three compositions, namely $Ni_{2.16}Mn_{0.84}Ga$, $Ni_{2.19}Mn_{0.81}Ga$ and $Ni_{2.20}Mn_{0.80}Ga$. In the $Ni_{2.16}Mn_{0.84}Ga$ sample the Curie temperature was well separated from the temperatures of structural transformation, so that austenite – martensite transition took place in a sample being in a ferromagnetic state [11]. As shown in Fig. 1, the structural transformation in this case was accompanied by a significant increase in sample length, but it was virtually non-sensitive to the external magnetic field.

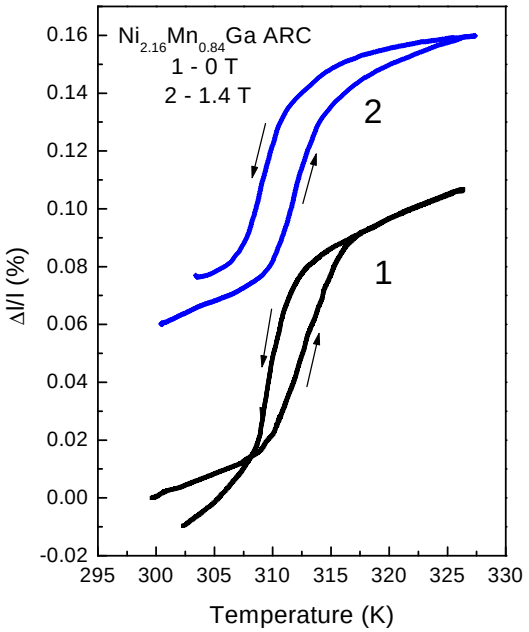

FIG. 1: Dilatometric effects in zero and 1.4 T magnetic fields observed in the $Ni_{2.16}Mn_{0.84}Ga$ polycrystalline sample.

In the $Ni_{2.19}Mn_{0.81}Ga$ and $Ni_{2.20}Mn_{0.80}Ga$ samples a different picture at the phase transition was seen. For these compositions the paramagnetic austenite transforms into ferromagnetic martensite [11]. The transformation upon heating is accompanied by a shortening of the sample. The change in the sample length at phase transition was 0.04% for $Ni_{2.19}Mn_{0.81}Ga$ and 0.12% for $Ni_{2.20}Mn_{0.80}Ga$. The same measurements performed in the presence of the 1.4 T magnetic field oriented perpendicular to the long axis of the sample showed a significant increase of the dilatometric effect of transformation. In the $Ni_{2.19}Mn_{0.81}Ga$ sample the effect of transformation increases by 3.2 times, i.e. from 0.04% to 0.13%, in the $Ni_{2.20}Mn_{0.80}Ga$ it increases by 2.6 times, i.e. from 0.12% to 0.31%. The increase of the dilatometric effect of transformation is seen also at cooling. The upward shift of

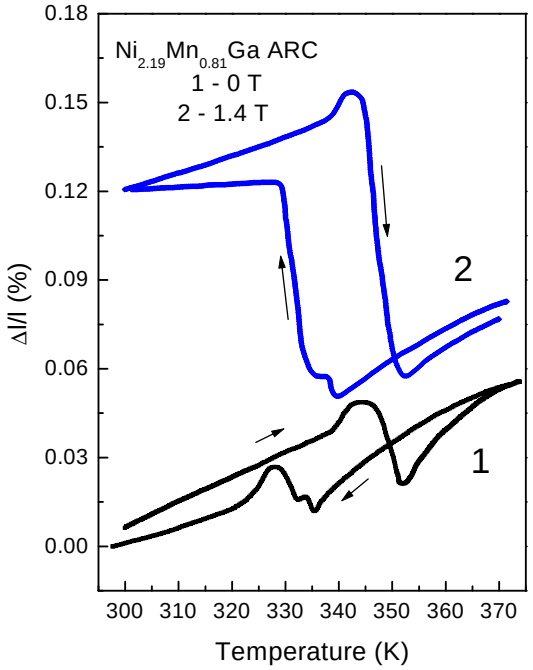

FIG. 2: Dilatometric effects in zero and 1.4 T magnetic fields observed in the $Ni_{2.19}Mn_{0.81}Ga$ polycrystalline sample.

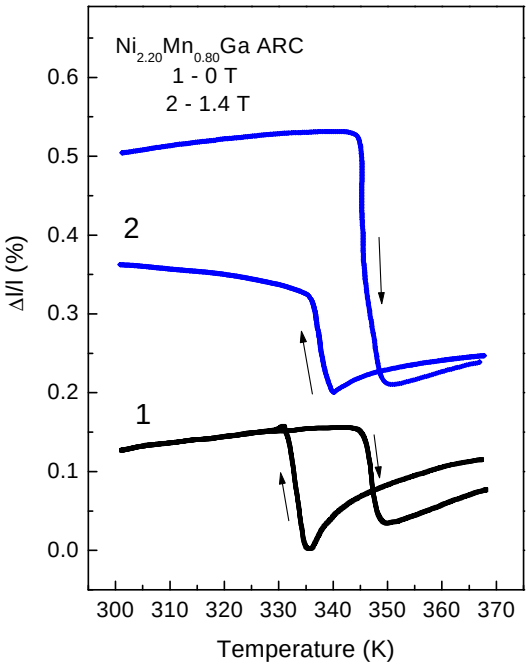

FIG. 3: Dilatometric effects in zero and 1.4 T magnetic fields observed in the $Ni_{2.20}Mn_{0.80}Ga$ polycrystalline sample.

transition temperature due to the external 1.4 T magnetic field is 1 K for $Ni_{2.19}Mn_{0.81}Ga$ sample and 2.7 K for $Ni_{2.20}Mn_{0.80}Ga$ sample. The results of these measurements are shown in Figs. 2 and 3.

The single crystal of $Ni_{2.19}Mn_{0.81}Ga$ was grown from the melt by the Czochralski technique. The dimensions of the as-grown sample were about 80 mm in length and about 12 mm in diameter. No special thermal treatment was employed to the crystal which showed clear martensitic patterns on its surface at room temperature. The crystal growth direction was [110]. The specimens for dilatometric studies were spark-cut from the large single crystal and had dimensions $3 \times 3 \times 6$ mm, where the longest dimension coincided with the crystal growth direction. A non-magnetic strain gage was attached along the crystal growth direction and the sample was inserted into the variable temperature chamber of a superconducting magnet. The phase transitions were easily detectable by the sharp changes in strain gage response. Since the phase transition in the sample studied is a first order it could be characterized by A_s and A_f, i.e. austenite start and austenite finish temperatures at heating, which were found to be 325 K and 333.5 K, respectively. At cooling, the M_s and M_f, i.e. martensite start and martensite finish temperatures, were found to be 329 K and 320.5 K, respectively. These data are presented in Fig. 4. In the absence of a magnetic field the sample exhibited elongation due to the thermal expansion in the martensitic phase with the rate of about 12.5×10^{-6}/K. In the vicinity of phase transition it exhibited a complicated behavior. Firstly, it shrinks by -6×10^{-4} in the range of 325 – 330 K, then elongates by 12×10^{-4} in the range of 330 – 333.5 K and finally exhibited elongation due to the thermal expansion in the austenitic phase with the rate of about 5×10^{-6}/K.

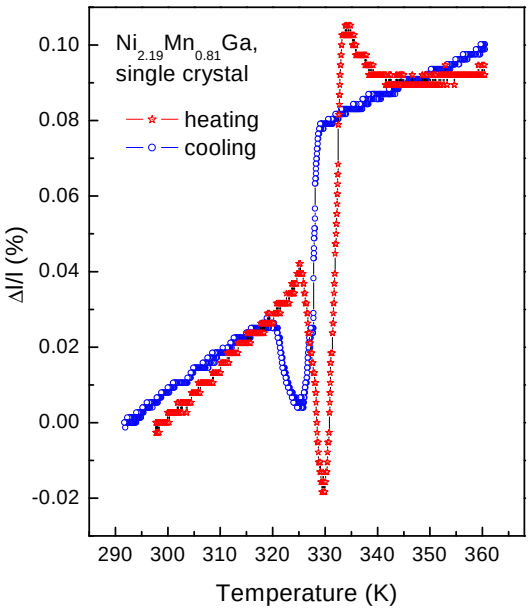

FIG. 4: Zero field strain in the $Ni_{2.19}Mn_{0.81}Ga$ single crystal upon heating and cooling.

The application of a 5 T magnetic field along the largest dimension of a sample resulted in the 5 K upward

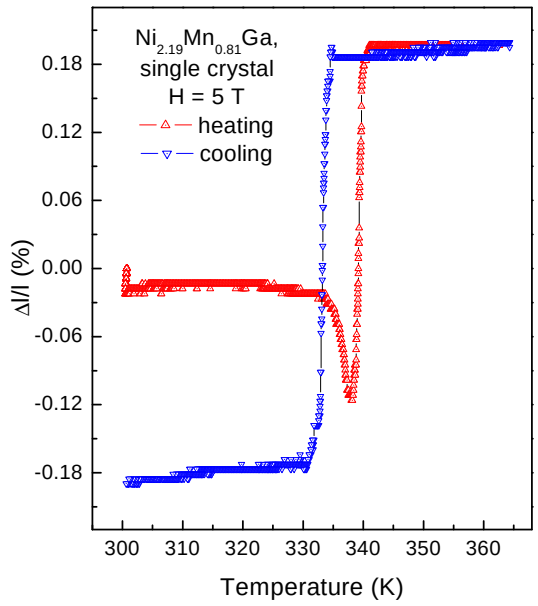

FIG. 5: Strain associated with the phase transition in $Ni_{2.19}Mn_{0.81}Ga$ single crystal in magnetic field 5 T.

shift of the critical temperatures. At 300 K the sample exhibited negative magnetostriction of about 4.5×10^{-4} at saturating field. At heating through the phase transition the sample shortens initially by 0.09%, then sharply elongates by 0.31%, afterwards it showed no temperature variation. At subsequent cooling, the sample showed a step-like downward change of about 0.4%. These data are shown in Fig. 5.

III. CONCLUSION

In conclusion, it was shown that in the temperature interval of the martensite – austenite transformation the ferromagnetic $Ni_{2+x}Mn_{1-x}Ga$ alloys show the giant magnetostrictive effects comparable with that observed in Terfenol-D. Taking into account the narrowness of the transition temperature range these observations open the possibility to induce complete phase transformation by the application of a magnetic field and to reach giant reversible magnetostrictive effect in the two-way trained shape memory alloys.

Acknowledgements

This work was partially supported by RFBR Grant No. 99-02-18247, by the Grant-in-Aid for Scientific Research (C) No. 11695038 from the Japan Society of the Promotion of Science and by the "University of Russia" Program.

[1] T. Shimozono, Y. Kohno, H. Kohishi et al., Effects of prestrain, heat treatments and magnetic field on martensite formation in Fe-25.5%Ni-3.5%Cr alloys, *Mater. Sci. Eng. A* **273-275** (1999), 337 - 341.

[2] H. Ohtsuka, G. Ghosh and H. Wada, Size and aspect ratio of martensite in Fe-Ni-C alloys formed under applied magnetic field and tensile elastic stress at 4.2 K, *Mater. Sci. Eng. A* **273-275** (1999), 342 - 346.

[3] T. Kakeshita, K. Shimizu, T. Maki et al., Magnetoelastic martensitic transformation in an ausaged Fe-Ni-Co-Ti alloy, *Scripta Metal.* **19** (1985), 973 - 976.

[4] R.D. James and M. Wuttig, Magnetostriction of martensite, *Phil. Mag. A* **77** (1998), 1273 - 1299.

[5] T. Kakeshita, K. Shimizu, S. Funada and M. Date, Magnetic field-induced martensitic transformations in disordered and ordered Fe-Pt alloys, *Trans. JIM* **25** (1984), 837 - 844.

[6] R. Tickle, R.D. James, T. Shield, M. Wuttig and V. Kokorin, Ferromagnetic shape memory in the NiMnGa system, *IEEE Trans. Magn.* **35** (1999), 4301 - 4310.

[7] G.H. Wu, C.H. Yu, L.Q. Meng et al., Giant magnetic-field-induced strains in Heusler alloy NiMnGa with modified composition, *Appl. Phys. Lett.* **75** (1999), 2990 - 2992.

[8] R.C. O'Handley, S.J. Murray, M. Marioni et al., Phenomenology of giant magnetic-field-induced strain in ferromagnetic shape-memory materials, *J. Appl. Phys.* **87** (2000), 4712 - 4717.

[9] H.D. Chopra, Ch. Ji and V.V. Kokorin, Magnetic-field-induced twin boundary motion in magnetic shape-memory alloys, *Phys. Rev. B* **61** (2000), R14913 - R14915.

[10] P.J. Webster, K.R.A. Ziebeck, S.L. Town and M.S. Peak, Magnetic order and phase transition in Ni_2MnGa, *Phil. Mag. B* **49** (1984), 295 - 310.

[11] A.N. Vasil'ev, A.D. Bozhko, V.V. Khovailo et al., Structural and magnetic phase transitions in shape-memory alloys $Ni_{2+x}Mn_{1-x}Ga$, *Phys. Rev. B* **59** (1999), 1113 - 1120.